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Literary review and modeling outlook for radiation damage in fused-silica optical fibers

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Abstract

The purpose of this document is to distill, from literature, the effects of radiation damage in fused-silica optical fibers. This information will be used to design a theoretical framework capable of predictive modeling that will be published elsewhere. By inspection of the relevant literature, the devised method is calculation of defect amorphous structures by molecular dynamic techniques, and computation of the macroscopic dielectric function using density functional theory. The dielectric function is then used to directly compute the transmittance of the core material before and after irradiation via solution of Maxwell's equations in a homogeneous medium.

1 Introduction

Optical fibers are primarily drawn glass used to transmit light. They are invaluable across space, nuclear, high-energy, and telecommunications applications. Sensors, data links, laser technologies, and others [1], all make use of optical fibers. Many of these applications demand that fibers endure harsh or extreme environments, especially, temperature and radiation. For example, space applications like satellites require optical fiber data links [2, 3], but operating outside the Earth's atmosphere exposes the fiber to large temperature gradients [4] and increased doses of radiation [5]. Nuclear and fusion facilities may need to couple light out of critical areas [6], and the proximity to nuclear reactions exposes the fibers bombardment by highly energetic particles. The safety of nuclear reactor vessels and nuclear power plants relies on monitoring systems that employ fiber optics [7]. Much recent effort has been devoted by Cheymol and coworkers [8] to test the effects of neutron irradiation on single-mode optical fibers. Facilities such as the Large Hadron Collider require sensing measurements that are often coupled out into optical fibers, and work has been done to enable radiation detection via the sensitivity of optical fibers to radiation [9]. In light of these applications, a non-empirical modeling scheme capable of predicting the optical/mechanical properties of fibers in harsh environments is greatly desired.

1.1 Summary of radiation damage effects

The modern understanding of radiation-induced damage in optical fibers is summarized in a 2020 report by Campanella et al [10]. They discuss the large-scale modeling effort to build CERTYF (Combined Effects of Radiations, Temperature and hYdrogen on silica-based optical Fibers), and lay out the central features of radiation-induced damage in optical fibers. It can be broken down into three parts.

1. Radiation-induced attenuation (RIA): a wavelength and time-dependent phenomenon corresponding to a decrease in the transmitted signal.
2. Radiation-induced emission (RIE): this consists of Cerenkov light or light emitted from some pre-existing or radiation-induced defects excited by the radiation.
3. Radiation-induced refractive index change (RIRIC).

An important comment made by Campanella et al [10] is: "***At the moment, a predictive model of the RIA, given the composition of the fiber and its environmental conditions, does not exist,***

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even if some semi-empirical models exist at fixed temperature of irradiation". They go on to say that the only current method of fiber qualification is through direct experimental characterization of its vulnerability. This means there is an obvious, current need in this field, and suggests any work towards a truly predictive modeling capability will have lasting scientific impact.

1.2 Goal of modeling effort and approach

Radiation-induced attenuation (RIA) is the most dominant effect, and is fundamentally a result of changes in the behavior of the light-matter interaction. This is captured by the macroscopic dielectric function ϵ ($\mathbf{D} = \epsilon\mathbf{E}$). The refractive index, emission, and absorption are all encapsulated in the macroscopic dielectric function. Understanding the optical effects of radiation then is reduced to being able to predict the dielectric function for a given material system. The material system before and after irradiation can effectively be viewed as a different material with different defect concentration and electronic structure. Therefore, the following question guides the focus of this review: ***What changes are induced in the opto-mechanical properties of silica fibers as a function of dose, dose rate, and temperature?***

2 Literature Review

2.1 Fundamental physics

This section reviews the fundamental physics of optical fibers and establishes a working knowledge of the literature in this regard – emphasis is placed on radiation resilience.

An optical fiber uses the index of refraction difference between the core and the cladding layer to instantiate a waveguide for light. If the dielectric properties of either medium are affected by exposure to radiation or temperature, the possibility of failure increases. Therefore, a lot of work has been devoted to understanding how the core's optical properties change as a function of radiation dose, dose rate, and temperature [11]. However, its interaction with the cladding is also important. See Figure 1 for a schematic representation of these components. We will focus on the core material, but some consideration may be given to the cladding layer. No consideration is given at this time to the materials outside of these layers.

For an in depth discussions regarding the fundamental, physical phenomenon that govern radiation-induced attenuation, Alessi et al [12] provide a general review of the importance of point defects in silica-based materials. They discuss the temperature dependence of defect diffusion as well as electron trap dynamics, and attribute most of the temperature dependence of RIA to these phenomena. De Michele et al [13] discuss origin of RIA in the presence of X-ray irradiation, and attribute RIA to self-trapped hole defects. In a 2019 work, Akchurin et al [14] discuss the radiation hardness in cerium-doped fused-silica optical fibers. This paper has several good references about fundamental theory. They find aluminum or cerium tend to make fibers radiation soft, and that a rate equation approach works reasonably well in the regime of 10-100 Gy and 10-100 mins, but 1-10 Gy/hr for months needs more work. In line with Campanella et al [10], Akchurin et al claim **"*molecular level simulations to investigate radiation effects in doped fused-silica are not yet available*"**, suggesting an atomistic approach would be valued by the community working on rad-hard fibers. S. Girard has led numerous investigations of irradiated optical fibers including several general purpose reviews [15–18], one review on space applications [19], two articles describing hole effects [20, 21], one article describing kinetics [22], and one article discussing neutron sensitivity [23]. ***Girard's exhaustive work shows the macroscopic responses are well described by semi-empirical modeling techniques, but a microscopic theory linking fiber chemistry with performance although is absent and highly desired.*** Finally, in a 1993 review by Griffioen et al [24], it is shown that of the 10 available lifetime models for fibers, they can all be reduced to 1 basic model. This work indicates that the fundamental issues regarding lifetime of fibers has been well described at the macroscopic level for many years, but in light of the more recent works calling for atomistic modeling, it provides more evidence that an atomistic approach will be a meaningful scientific route to follow.

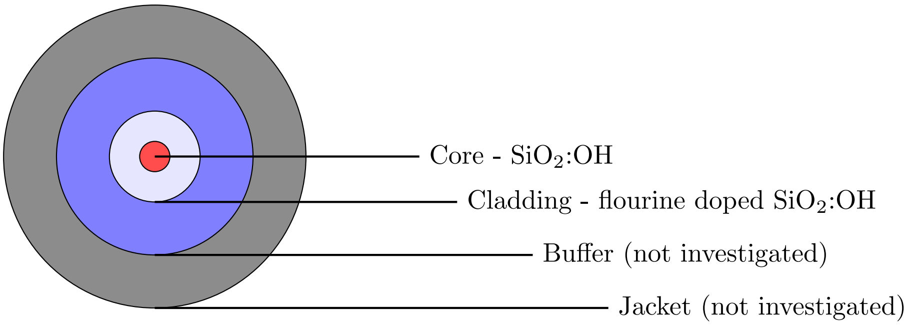

Figure 1: Schematic of typical fiber construction

2.2 Temperature effects

Temperature affects the lifetime of radiation-induced defects, which in turn, affects the absorption characteristics of fibers. Kuhnhenn et al [25] review the temperature effects in irradiated fibers. ***In general, radiation-induced damage effects decrease with increasing temperature***. This behavior is logical because increased thermal energy will allow more defects to spontaneously annihilate.[1] There is however literature to suggest this general trend is wavelength and composition dependent. For example, Jin et al [27, 28] comment on the temperature dependence of RIA in the near infrared (NIR), and find the usual inverse relationship between RIA and temperature does not hold in the NIR. In this case, the RIA is decreased at lower temperatures, contradicting the notion that thermal energy helps anneal radiation-induced defect centers. Additional work by Jin et al [29] highlights several key factors of the temperature dependence of RIA

- The temperature dependence of RIA in an irradiated fiber is wavelength-dependent.
- The RIA in fibers with a higher total radiation dose is more sensitive to temperature.
- The transmission wavelength has the maximum impact on temperature dependence of RIA

Their data does show that during the first 5000 seconds at room temperature, the loss decreases, however, as the temperature is elevated to 60 °C over 15000 seconds, the loss increases. The fact that this result is wavelength dependent aligns with the notion that thermal energy enables the creation/annihilation of a particular species of defect, and even at equilibrium, specific defects retain a significant population. Additional evidence that operating wavelength and composition affects RIA is given by Vecchi et al [30]. Alessi et al [31–33] measure the radiation-induced attenuation (RIA) in Ge-doped optical fibers and find that the temperature dependence of RIA depends on the dose. The general trend however is the RIA is a linearly decreasing function of excitation wavelength. Different Ge defect types available at different temperatures are attributed to the different RIA dependencies as a function of temperature. Blanc et al [34] measure RIA at cryogenic temperatures in regards to applications at the Large Hadron Collider (LHC). They claim RIA inversely varies with temperature because annealing can almost completely heal any radiation damage. At cryogenic temperature, no annealing can take place, and there is a huge RIA.

2.3 Rate equation modeling

The dependence of radiation-induced induced attenuation generally follows a power law function of the dose. This is an experimental observation. Significant effort has been devoted to understanding the origin of this power law, as well as developing predictive models that leverage it. This statement is discussed theoretically in this section, and its experimental counterpart is discussed later in section 2.5. The Griscom et al paper [35] is a fundamental result in this regard. It is tour de force of radiation effects prediction. The observed universal behavior of tens of thousands of condensed matter samples all showing the time dependence of a relaxation process $q(t)$ going as

$$q(t) = q_0 \exp\left[-\left(\frac{t}{\tau}\right)^{\alpha}\right] \tag{1}$$

[1] This is in line with previous work showing the transmission characteristics of neutron irradiated fibers returned to their pristine levels within one minute at room temperature [26].

is discussed as originating in (i) stochastic diffusion-controlled reactions in disordered solids and (ii) "hierarchically limited dynamics resulting in correlated relaxation processes consisting of several successive steps". This motivation allows them to conclude, after direction validation of the concept against experiment, that the experimental power-law function for growth kinetics is actually the envelope function of a series of first-order rate equation solutions each with a unique production rate K_i and recombination rate R_i. Along these lines, Borgermans and Brichard [36] investigate the spectral and kinetic behavior of gamma irradiated pure silica fibers in spent fuel facilities. They develop a simple model $\dot{n} = a\dot{D} - n/\tau$ where n is defect concentration, $\dot{D}$ is the dose rate, τ is the lifetime. The important aspect is both dose and temperature come into play even at level of first-order kinetic equations, and attenuation (A) follows power law functions of dose (D) as

$$A = CD^f, \tag{2}$$

where C and f are unknown constants. Gilard et al [37] work along similar lines with an n^{th}-order rate equation model to describe the dose, dose rate, and temperature dependence of RIA in fibers. This works well to fit power law dependencies, but at this time, its advantage is not clear. Liu and Johnston [38] build a theory following arguments similar to Griscom [35]. It is based on super-posing the individual generation and decay of optically active defect centers, but the dose-rate dependence can be incorporated. They focus on the recovery time, and find good agreement between theory and experiment. Recent simulation work was performed by Ma et al [39]. They build a rate equation analysis and are able to investigate radiation effects. The role of defect production can be treated within a rate equation approach [40].

2.4 Atomistic modeling

Here we review theoretical works targeting first-principles modeling. The 2018 book by Bagatin and Geradin [41] demonstrates the need for a multi-physics approach spanning multiple length and time scales. It shows that there are current efforts to design a workflow starting with molecular dynamics, moving through density functional theory, and using advanced techniques like solution of the Bethe-Salpeter equation to understand the optical properties of irradiated glasses. The use of GW and BSE methods for a-SiO_2 suggests that complex many-body effects may play a significant role in these materials as well. We briefly review each step in this workflow in turn.

Molecular dynamics of SiO_2 is studied rather extensively. A 2020 report by Okada et al [42] has many useful references regarding the molecular dynamics of irradiated silica. Next, connecting the defect wave functions to the macroscopic optical properties comes through the dielectric function $\epsilon(\omega)$. This is available with modern density functional theory [43, 44] (although still non-trivial). Finally, we seek to know the change in transmittance. Fugallo et al [45] show how to get transmittance from dielectric function $\epsilon(\omega)$. The reflectivity r and power loss factor τ are defined as

$$\tau = \exp\left[-4\pi d \cdot \mathrm{Im}\left(\frac{\sqrt{\epsilon}}{\lambda}\right)\right], \quad r = \left|\frac{\sqrt{\epsilon}-1}{\sqrt{\epsilon}+1}\right|^2, \tag{3}$$

with the transmittance $\mathcal{T}$ as

$$\mathcal{T} = \frac{\tau(1-r)^2}{1-r^2\tau^2}. \tag{4}$$

These equations give a transparent connection between DFT and the measurements. Because the primary microscopic effect of radiation damage is creation of defects, it is important to investigate the energetics of defects in crystalline SiO_2. Work along these lines has been done by Yue et al [46] and Giacomazzi et al [47].

Several works at the level of DFT+GW+BSE have also been done for advanced modeling of optical properties [48, 49]. These works discuss optical properties of defects, photobleaching, generation of defects during drawing out of optical fibers, and the role of exciton physics. There are some exciton states ~ 1 eV below the fundamental gap ~ 9 eV. El-Sayed et al [50, 51] find evidence for electron/hole localization in a-SiO_2. Localization centers are created, new absorption bands are created, and annealing can remove the absorption bands. They additionally discuss defect statistics in a-SiO_2.

2.5 Experimental characterization

This section comments on experiments investigating RIA. In general, γ-rays and X-rays have the same qualitative effect, but the γ-ray effects are more pronounced. Neutron damage produces oxygen vacancy type defects, while photon irradiation instantiates oxidation/reduction processes.

The types of defects created by radiation exposure depends on the initial fiber composition. Wen et al [52] and Devine et al [53] perform spectroscopic investigation of defects by Raman and EPR. Blanchet et al [54] show that proton and X-ray irradiation induce similar effects at 10kGy, but electron beam irradiation effects depend on fiber composition. B/Ge rich fibers are sensitive to electrons, H_2-loaded P/Ce fibers are susceptible to X-rays. This means the sensitivity to a particular radiation source is linked to fiber composition. Thus, it is desirable to measure the relation between composition and radiation-resilience. Wen et al [55] and Reghioua et al [56] show that the existence and impact of point defects in silica-based fibers can be effectively investigated with luminescence measurements.

Along similar lines, Al-Helou et al [57] measure optical absorption and photoluminescence intensities of Cu doped SiO_2 before and after X and γ irradiation. They find radiation-induced absorption bands, and infer change in Cu^+ local symmetry. Kim et al [58] find the Verdet constant of Cu-doped germano-silicate fibers to increase 1.46 fold post γ-irradiation. This means the birefringence of the material has been enhanced.[2] The effects of x-rays have been studied by De Michele et al [59]. They characterize canonical optical fibers with different dopants at different temperatures, and claim RIA in visible spectrum well understood, while RIA in the infrared is less understood. Recent experimental data is in line with historical data [60].

Skuja et al [61] discuss neutron-irradiated α-quartz (SiO_2) crystal and show non-bridging oxygen hole centers (oxygen dangling bonds) contribute to photoluminescence at 20 K, but also to crystal-like zero-phonon line excitations. Contrasting neutron and photon irradiation, neutron shows unique effects suspected to come from direct modification of the silica network [62], and that only neutrons are capable of inducing oxygen defect centers [63]. Skuja et al [61, 64] show that neutron irradiation can amorphize parts of crystalline SiO_2. They also show that phonon excitations become important in the presence of silicon dangling bonds. This 2019 work by Petrie et al [65] investigates high-dose neutron damage both experimentally and theoretically. They show hydrogen loading can improve radiation-resilience in the UV and visible ranges, but leads to increased absorption in the IR range.

Finally, Kashaykin et al [66, 67] discuss the role of strain and temperature in the formation of self-trapped holes (STH), and how these play into the radiation-induced attenuation. The conclusion is that multiple types of defects are possible, but one type is favored in strained samples and another type is favored in native samples. On the other hand, strained samples can have long live STH's, and therefore enhanced RIA. Piccolo et al show that 1% strain can be significant [68]. The fiber drawing temperature (intrinsic strain) is shown to have significant effect on the radiation-resilience of fibers [69]. ***All this work suggests that the transmittance profile will be different for the strained and unstrained fibers.***

3 Summary

The use of optical fibers in radiation-extreme environments is increasing, yet fundamental understanding of damage mechanisms and associated quantitative modeling toolsets are far from complete. Because the ability of a fiber to transmit light degrades with radiation-induced material point defects, an effective modeling and simulation capability requires bridging length scales from nanometer to kilometer. The literature reviewed in this document suggests the best approach will incorporate a multi-physics method comprised of classical molecular dynamic treatment of atomic defects, a quantum treatment of the light-matter interaction, and a classical solution of Maxwell's equations for light inside a damaged fiber.

[2] The ability of left and right circularly polarized light to interact with the material has been altered.

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